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Rend 8/21/02

Dear Dr Masten,

Re Federal Register (June 12, 2002; Vol. 67, No 113, pages 40329-40333.) notice for Public Comments, Hexafluorosilicic acid [16961-83-4] and Sodium hexafluorosilicate [16893-85-9].

Before responding to the National Institute of Environment and Health Sciences' Request for Public Comments, I would make the following preliminary statement:

The EPA and NIEHS acknowledge that the chemicals mentioned above have been used for decades to artificially fluoridate the drinking water supplied to 180 million Americans, but they admit to having no expert knowledge of the pharmacokinetics and toxicokinetics of the products in question.

In discussions on the problems of identifying the fluorosilicate radical in aqueous solution with a number of manufacturers of 'state-of-the-art' analysis equipment, all expressed confidence that it exists, although their equipment is unable to identify it at this time.

The NIEHS nomination stated: "Toxicological studies may be considered when results of chemical characterization studies are available for review". At first glance, this caveat appears to present a very high hurdle indeed, which might be seen to provide a loophole by which the USEPA and the NIEHS could avoid recommending research conducted with either of the fluoridation agents.

As will be shown, the "characterization" criteria were met by Bauman in 1947.

Therefore, the Government agencies should act immediately by requesting the National Toxicology Program to determine the safety of the fluoridation agents.

## **OBSERVATIONS**

The NIEHS/NTP, EPA and Centers for Disease Control have consistently denied that fluoride speciation occurs in drinking water. They have not exhibited any interest in the toxicological implications of fluoride species added to community drinking waters, nor those which may be created during the water treatment process, e.g., fluoroaluminum complexes. Indeed, the agencies continue to assure citizens that speciation does not occur. See, for example, the following excerpt from a recent CDC email to a member of the public:

Added to the drinking water in very small amounts, the fluoride chemicals dissociate virtually 100% into their various components (ions) and are very stable, safe, and non-toxic.

We, the scientific community, do not study health effects of concentrated chemicals as put into water, we study the health effects of the treated water, i.e., what those chemicals become: fluoride ion, silicates and the hydrogen ion. The health effects of fluoride have been analyzed by literally thousands of studies over 50 years and have been found to be safe and effective in reducing tooth decay. (email from <a href="mailto:nccdohinfo@cdc.gov">nccdohinfo@cdc.gov</a> CDC Division of Oral Health, July 17, 2002, 10.05 AM.).

The Oral Health information department of the CDC apparently has no knowledge of a paper dated June, 2000, in which Urbansky and Schock (USEPA) wrote:

There are many metal cations competing for the fluoride; therefore, the free fluoride available to complex with the lead (II) ion is very small. In addition, most, if not all, of the competing metal cations are in greater abundance than the lead (II) by orders of magnitude . . . That drinking water contains a substantial fraction of fluoroaluminum complexes rather than free fluoride was highlighted by Pitter [1985] as a concern because free fluoride is more effective in protecting against tooth decay."

The Oral Health information department is also in the dark about a report, dated October 2000, entitled "Aluminum Compounds: Review of Toxicological Literature, Abridged Final Report." This Review, also prepared for Scott Masten, PhD, accorded substantial space to both speciation and the potential neurological effects of aluminum/fluoride in drinking water.<sup>2</sup>

However, the findings reported in June and October 2000 were certainly not new.

## **BACKGROUND**

In "Fluoride Drinking Waters" (Ed. F.J. McClure, 1962, USPHS), speciation of fluoride in drinking water is discussed in several articles and studies, e.g., "Fluoridation of Public Drinking Water", F.J. Maier, pp. 258-263. The section on "Side Effects," (page 261), is noteworthy in the context of the NIEHS "characterization" criteria:

Fluoridation will provide all or part of the fluorides needed to remove silica from boiler water. An ion exchange process is reported to require 1.0 ppm fluoride for each 0.5 ppm silica removed. On the other hand, if sodium silicofluoride is used for

fluoridation, about 0.5 ppm silica is formed when 1.0 ppm is added from this source. (Bauman 1947, cited by Maier.)<sup>3</sup>

We can see from Bauman's experimental findings, which were acknowledged and accepted by Maier and McClure, that (a) the fluorosilicate radical *does* exist in some form in water, and, (b) that complete dissociation does not occur.

Therefore, the NIEHS "characterization" criteria were met by Bauman more than 55 years ago!

## DISCUSSION

The NIEHS Review paid scant attention to the charts published by Kick and McClure. This is evidenced by the reviewers' conclusions on the differences in fluorine absorption and retention rates. On the last page the reviewers combine the work of Kick, et al and McClure in the same paragraph. Of Kick's work, the NIEHS Review states:

In a comparative study of absorption and excretion of fluorine in rats fed sodium fluoride, calcium fluoride, and sodium hexafluorosilicate, the percent fluorine retained was the same for the two sodium compounds (Kick et al., 1935 [see Section 9.1.2 for details regarding sodium hexafluorosilicate]).

See Table below:

Kick et al 1935 no 61

Fluorine Supplement	Time in Days	Fluorine ingested	Fluorine in feces	Fluorine absorbed	Fluorine in urine	Fluorine balance	Fluorine retained
		Mg.	Mg.	Mg.	Mg.	Mg.	%
Rock Phosphate	10	213.6	131.5	82.1	20.5	+61.6	28.8
Sodium Fluorsilicate	23	269.9	94.3	175.6	93.6	+82.0	30.4
Sodium Fluorsilicate	22	269.9	94.4	175.5	90.2	+85.3	31.6
Sodium Fluoride	18	211.2	116.5	94.7	25.8	+68.9	32.6
Calcium Fluoride	11	229.6	225.5	4.1	4.2	-00.1	0.0

It can be readily observed, from this experiment with rats, that

- 1) The amount of <u>fluorine found in feces</u> in the sodium fluorosilicate group was 94.4 mg. The result from the sodium fluoride group was 116.5 mg. *Note the difference*.
- 2) In the urine of the sodium fluorosilicate group, the amount of fluorine found was 90.2 mg, while 25.8 mg was found in the sodium fluoride group. Note the difference.
- 3) The amount of <u>fluorine absorbed</u> by the sodium fluorosilicate group was 175.5 mg. From the sodium fluoride group it was 94.7 mg. Note the difference.
- 4) The <u>fluorine retained</u> from the sodium fluorosilicate group was 31.60 mg and from the sodium fluoride group it was 32.62 mg. *Note the difference*.

The bioavailability of the fluorosilicates is much higher in the first three examples. In example 4, whether the retained fluorine is from sodium fluoride or from sodium fluorosilicate, 1.02 mg is a very large difference in a juvenile rat! It should be remembered that 1mg/F added to one liter of drinking water is intended to create physiological change in human youngsters!

These differences warrant urgent toxicological investigation and explanation.

Commenting on the McClure study, the NIEHS Review states:

Several experiments on growing rats orally given 5, 10, 15, 25, and 50 ppm fluorine as sodium fluoride or sodium hexafluorosilicate for 90-100 days found no differences in the quantity of fluorine deposited and the contents of ash, calcium, and phosphorus in the incisor teeth, molar teeth, mandibles, and femurs. Furthermore, there were no differences in the percent of ingested fluorine retained in the body, and a combination of sodium silicate (15 ppm silicon) with sodium fluoride (25 ppm fluorine) did not affect the amount of fluorine deposited. The growth rate was normal in all rats (McClure, 1950).

McClure's tables (published 1950) show clear differences in the average daily weight gains between the sodium fluoride and sodium fluorosilicate groups. For example:

Table 2 shows the average daily weight gain of rats consuming 5 mg/F per day. The sodium fluoride group gained 1.89 gm. In the sodium fluorosilicate group the gain was 1.76 gm.

Paradoxically, Table 4, on rats consuming 25 mg/F per day showed a reverse effect, i.e. rats on sodium fluoride gained 1.29 gm per day, while those on sodium fluorosilicate gained 1.52 gm.

These differences were either not noted, or they were missed by the authors of the NIEHS Review, but they must be taken into consideration.

The NIEHS reviewers relied heavily on two studies by McClure. Apart from noting the weights of rats, these studies were mainly concerned to determine fluorine uptake in bone and teeth. They were not toxicological studies.

Throughout McClure's work on fluorosilicates, it was evident that his purpose was to find the least expensive means to artificially fluoridate drinking water. Here are extracts from the opening paragraph of each of his studies:

- Thus with sodium fluorosilicate currently selling at about half the price as sodium fluoride, the cost of chemicals for fluoridation of 1 million gallons of water at the optimum level of 1.0 p.p.m. fluorine is approximately \$2.12 using sodium fluoride and 76 cents for an equivalent quantify of sodium fluorosilicate. (McClure, 1950)<sup>4</sup>
- Although Sodium fluoride (NaF) is now the most common fluoride in use for community water fluoridation, other compounds particularly sodium fluorosilicate (Na2SiF6), if found to be comparable to NaF in physiological effects\*, may have an advantage in being produced at less expense than NaF. Since commercial grade NaF is 95% pure, and at current market quotations sells for 11 cents per pound, and commercial grade Na2SiF6 is 98% pure and sells for 5 cents per pound, the cost per pound for available fluoride is approximately three times more for NaF than Na2SiF6. (Zipkin, McClure, 1951).5

<sup>\*</sup> Twenty years earlier, McClure had already established that there were differences in the physiological effects. (McClure, Mitchell, 1931).

Although sodium fluorosilicate, Na2SiF6, is cheaper than sodium fluoride, NaF, its use
for fluoridation of municipal water supplies may be limited to some extent by its solubility.
Ammonium fluorosilicate, (NH)2SiF6 is considerably more soluble than either of these
two fluorine compounds, and it is cheaper than NaF. (Zipkin, McClure, 1954).<sup>6</sup>

No consideration for health and safety factors is noted in McClure's work. However, he did know, from his early animal nutrition work, (McClure Mitchell, 1931) that toxicokinetic/pharmacokinetic differences existed between sodium fluoride and fluorosilicates. He neither followed through with toxicological studies himself, nor did he recommend any.

[Although these studies on the bioavailability of fluorine in bones and teeth did not recommend further physiological investigations, they are widely cited as "proof" that artificial water fluoridation is "safe and effective." However, on close scrutiny, the studies appear to have been crafted to form part of a product sales pitch; the authors even listed the prices of the three compounds!]

## **CONCLUSION**

Artificial water fluoridation is enthusiastically recommended and promoted by the CDC/DHHS, the parent organisation of NIEHS. All of these agencies are aware that fluorosilicates have been added to the drinking water of more than 180 million Americans for several decades.

Bauman's 1947 experimental finding that the fluorosilicate radical exists in water was acknowledged and accepted by both Maier and McClure. 78

The documented physiological differences observed by Kick, et al and McClure, et al, strongly support the conclusion that all water fluoridation studies in which sodium fluoride was - and still is - used as a laboratory surrogate for fluorosilicates are invalid and should now be discarded.

Yours sincerely,

References

<sup>&</sup>lt;sup>1</sup> Urbansky, E.T., and Schock, M.R.. Can fluoridation affect water lead levels and lead neurotoxicity? In: American Water Works Association Annual Conference Proceedings, Denver, CO, June 11-15, 2000.

<sup>&</sup>lt;sup>2</sup> Aluminum Compounds - Review of Toxicological Literature, Abridged Final Report prepared for Scott Masten, Ph.D., National Institute of Environmental Health Sciences, Contract No. N01-ES-65402, Submitted by Bonnie L. Carson, M.S. Integrated Laboratory Systems, October 2000.

<sup>&</sup>lt;sup>3</sup> Bauman, et al, Silica Free Boiler Water by Ion Exchange, Ind. Eng. Chem. 39:1453, 1947.

<sup>&</sup>lt;sup>4</sup> McClure, F.J. Availability of fluorine in sodium fluoride vs. sodium fluosilicate. Pub. Health Rep. 65:1175-1186, 1950..

<sup>&</sup>lt;sup>5</sup> Zipkin, I., McClure, F.J; Complex Fluorides, Caries Reduction and Fluoride Retention in Bones and Teeth of White Rats. Pub Health Rep, 66: 1523-1532, 1951.

<sup>&</sup>lt;sup>6</sup> Zipkin, I., and McClure, F.J; Cariostatic effect and metabolism of ammonium fluosilicate. Pub. Health Rep. 69:730-733, 1954.

<sup>&</sup>lt;sup>7</sup> Maier, F.J; Fluoridation of Public Water Supplies. J. Am. Waterworks Assn. 42:1120-1132, 1950.

<sup>&</sup>lt;sup>8</sup> Fluoride Drinking Waters, F.J. McClure, published by US Public Health Service, 1962.